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The Geometric Area Shape Factors of Coals Ground in a Standard Hardgrove Mill

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INTRODUCTION

The relation of energy input on grinding to the production of fresh broken surface has received much study (1); and, consequently, comparative increases in surface areas of ground particles have been measured by a variety of methods. However, the absolute external or geometric surface area of coal particles, and the corresponding shape factor, have significance in other processes than grinding. For instance, in processes where coal is chemically reacted at high rates, the rate of reaction is partially controlled by the geometric area (2). The area of any sample of coal of which the density and sieve size distribution are known can be readily calculated from a knowledge of a shape factor k defined by

$$(\text{geometric surface area})\ \mu = k\ (\text{volume}) \qquad (1)$$

where μ is a sieve size. Aris (3) has shown that the ratio of volume to geometric area, that is μ/k, is a characteristic dimension for use in studies of the reaction of solids in beds. Hawksley (4) considers that the drag diameter, which is approximately equal to the diameter of the sphere having the same surface area as the particle, is the most fundamental dimension for use in hydrodynamic problems involving particles. The drag diameter is related to the sieve size by

$$d = \frac{6\mu}{k} \qquad (2)$$

The above remarks indicate that a knowledge of the values of k for coals is likely to be of interest in many industrial processes. Examples of such processes where the shape factor is of actual or potential importance are:

(a) the combustion of pulverized coal in central power stations,
(b) the gasification of pulverized coal,
(c) the production of low temperature coke and coal chemicals in fluidized bed processes,
(d) the firing of open cycle turbines with pulverized coal,
(e) the hydrogenation and chemical processing of coal suspensions, and
(f) the transport of coal in fluid suspensions.

REVIEW OF PREVIOUS WORK

As is well known, coal has a large internal surface area due to a very fine micropore system within the particle (5). Many methods of measurement of surface area determine this internal area in addition to the exterior

surface and, therefore, are not suitable for the measurement of shape factors. The fluid permeability technique, utilizing a modified Kozeny equation, does not in general measure internal surface area (6). This is so because the internal pore system of coal has a negligible permeability compared to the flow between coal particles in a packed bed. Romer (7) used an air permeability method to measure the geometric surface area of ground coal fractions. He did not, however, make available enough information to enable shape factors to be calculated. Skinner (8) pointed out that the hydrodynamic area as measured by permeability methods is of primary importance in the combustion of pulverized fuels and recommended that it should be used to measure shape factors instead of assuming a constant shape factor with size.

APPARATUS AND EXPERIMENTAL TECHNIQUE

Grinding of Coal--The coal under test was ground in a standard Hardgrove test machine according to the A.S.T.M. standard method (9). The ground product was carefully sieved into fractions and the fractions weighed. These fractions were then used for the density and surface area measurements described below.

Density of Coal Fractions--To measure the surface area by the permeability method, it was necessary to know the apparent density of the coal particles tested. Apparent densities were determined, at first, by water displacement in a specific gravity bottle. The coal powder was weighed, vigorously stirred with water to thoroughly wet and release entrapped air, and the density determined. The operation could be carried out in a few minutes and it was thought that in this time the water would not penetrate the internal porosity of the coal to any marked extent. This method gave reproducible results for the larger mesh fractions, but it was found difficult to wet and de-aerate the fine mesh fractions (-200 mesh). Consequently, mercury densities were determined using a mercury porosimeter (10). The pressure of mercury was increased until the rate of entry of mercury dropped suddenly and further pressure caused only a small further penetration. The sudden change-point was taken as equivalent to the geometric density, and the slow additional penetration as the filling of the internal pore system of the particles.

Measurement of Surface Area of Coal Fractions--The surface areas of the coal fractions were determined using the liquid permeability apparatus described by Lakhanpal, Anand and Puri (11) and shown in Figure 1. A coal sample was weighed and packed into tube A, being supported by a thin pad of glass wool over the constriction in the tube. The time taken for a given volume of water to flow through the bed under the mercury head was noted, as were the initial and final mercury heads, suitably corrected for the water pressure head on the right hand column of mercury, Δl. From the weight and density of the coal sample, the diameters of tubes A and C (determined by mercury calibration), the length of the bed, and the viscosity of water at the temperature of the experiment, the surface area of the coal in $cm.^2$ per g. of coal was calculated from equation (3a).

The method was found to be simple and quick and to give good reproducibility. The major difficulty was found in obtaining a bed free from air bubbles. When air was present, the bed had a characteristic mottled appearance at the surface of the tube and surface areas were both too high and poorly reproducible. This difficulty was overcome by allowing the coal

sample to soak in boiled-out water for an hour before use, with frequent stirring, and by packing the bed wet. If the coal was well wetted and packed under suction (from a water pump) with a continuous flow of boiled-out water, air bubbles were not found in the bed. The bed was kept completely full of air-free water at all times during the testing.

The instrument was tested by measuring the surface area of a sample of glass spheres of size 100-200 microns, the glass having a density of 2.50 g./cc. A microscope size count was made on the spheres and the surface area calculated as described later. Comparing the two areas gave a mean factor of k_0q^2 in equation (3) of 4.75, with the values from six tests lying always within ±6% of the mean. Carman (12) reviews values of k_0q^2 found for spheres and quotes values from 4.5 to 5.1, with a best value of 4.8. The value found was in good agreement with this, and it was concluded that the apparatus was working satisfactorily.

THEORY

Specific Surface Area of Tested Material--The specific surface area of a coal fraction was calculated from the following formulae, which can be easily derived.

$$d = 5\left(\frac{k_0q^2}{4}\right)^{\frac{1}{2}}\left(\frac{R_1}{R}\right)\left(\frac{1-\epsilon}{\epsilon}\right)\sqrt{\frac{\eta L \log(\ell_2/\ell_1)}{\epsilon t}\,10^5} \qquad (3)$$

$$S_0 = \left(\frac{6}{d}\right)10^4 \qquad (4)$$

where d is the surface area mean spherical diameter of the material in microns, S_0 is the specific surface area in sq. cm. per $cm.^3$ of material, k_0q^2 is a factor which varies with the shape of the pores in the bed, R is the radius of tube A, R_1 is the radius of tube C, ϵ is the porosity of the packed bed, η is the viscosity of water, L is the length of the packed bed, ℓ_1 and ℓ_2 are the pressure differentials across the bed initially and finally in cm. of mercury, and t is the time of flow in seconds. The factor k_0q^2 is 4 for circular pores, 4.8 for a bed composed of spheres and 5 for a bed composed of irregular particles (13). It should be noted that d is only an intermediate step in the calculation of the surface area S_0, and it is not necessary to attach any particular physical significance to it. However, values of d were calculated because these values could be compared with the nominal sieve sizes of the material tested. These values, consequently, gave a ready indication of an unsatisfactory test. S_0 is in no sense a mean of determined dimensions but is a direct measure of surface area. For the irregular particles of ground coal, equation (3) becomes

$$d = 5\sqrt{\frac{5}{4}}\left(\frac{R_1}{R}\right)\left(\frac{1-\epsilon}{\epsilon}\right)\sqrt{\frac{\eta L \log(\ell_2/\ell_1)}{\epsilon t}\,10^5} \qquad (3a)$$

In order to test the accuracy of the apparatus, the specific surface area of a sample of glass spheres was determined by microscope measurement and by the permeability method. The specific surface area by microscope measurement was calculated by using the following equation

$$S_o = 6 \int_0^N \mu_i^2 \, dN_i \Big/ \int_0^N \mu_i^3 \, dN_i \qquad (5)$$

where dN_i is the number fraction of spheres in the microscope diameter range $\mu_i + d\mu$. The integrations were performed graphically by plotting the cumulative number of particles below diameter μ_i against μ_i^2 and against μ_i^3 and finding the appropriate areas under the curves.

b) Calculation of area-to-volume shape factors.

The specific surface areas per $cm.^3$, S_o, of 10 sieve fractions of a coal ground according to the standard Hardgrove test (60 grinding revolutions) were determined. As the percentage weight of coal in any fraction, Δp say, and the density ρ were known, the surface area ΔS of the fraction was calculated from

$$\Delta S = \frac{S_o}{\rho} \cdot \Delta p$$

The cumulative total surface area, S, and cumulative weight p were then calculated from

$$S = \sum \frac{S_o \Delta p}{\rho}$$

$$p = \sum \Delta p$$

When S was plotted against p on semi-log paper, it was found that a smooth, shallow curve was obtained. Then

$$\frac{dS}{dp} = \frac{dS}{d(\log p)} \left(\frac{1}{2.3p} \right) \qquad (6)$$

and values of $dS/d(\log p)$ were obtained from the slope of the curve at any given value of p. The specific surface area per gram at a sieve size μ corresponding to weight p is clearly given by

$$S_\mu = \left(\frac{dS}{dp} \right)_\mu \quad cm.^2 \, g.^{-1} \qquad (7)$$

If ρ_μ is the density of coal of size μ and the shape factor for this size is k_μ from equations (7) and (2)

$$k_\mu = \left(\frac{dS}{dp} \right)_\mu \mu \rho_\mu \qquad (8)$$

It should be noted that k_μ is the shape factor at size μ and is not a mean over a range of sieve sizes. k_μ is independent of the size distribution of the ground coal, whereas any mean value of k would depend on the size distribution between the sieve sizes averaged.

RESULTS

Table 1 gives the analyses of the coals used in the tests. The four coals cover a range from low rank high volatile sub-bituminous coal to high rank low volatile anthracite.

Figure 2 gives the variation in apparent density of the ground coals with size. In general, the value of density determined at a given size varied about the mean line within a range of ±3%. Fortunately, the value of dS/dp in equation (6) is insensitive to small changes in density. The effect of density is mainly the direct proportionality shown in equation (8). The water densities,where determined, were equal to the mercury densities within the limits of accuracy of both methods.

Table 2 gives the size/weight distribution of the four coals ground for the standard 60 revolutions and the specific surface areas of the ground fractions. At least three area measurements were made on each fraction; the values obtained were always within ±3% of the mean and were usually within ±1%.

Figure 3 shows the cumulative surface area of material less than 30 mesh (U.S. standard sieve) plotted against the percentage weight undersize for coal B19426. Similar curves were obtained for the other coals tested. The numerical values of the slope were used to obtain shape factors using equation (6) and (8). Table 3 gives the shape factors at various micron sizes. Over the size range investigated (approximately 40 to 600 microns), the shape factor was found to be constant for a given coal, the determined values varying randomly about the mean within about ±4%. There were marked differences in the shape factors for the four coals investigated, the differences being considerably greater than can be explained on the basis of experimental errors of the various determinations made.

DISCUSSION OF RESULTS

The variation of apparent density of coal with size after grinding is to be expected, since stronger material will tend to collect in the coarser fractions. The stronger material will usually be denser since it will contain more mineral matter and denser, more coalified coal particles. This effect is more marked for the weaker coals for two reasons: a) there is a greater difference between the hardness of the mineral matter and the remainder of the coal and b) the grinding has proceeded to a more advanced stage for the weaker coals (although ground for the same number of revolutions). The concentration of denser material in the coarse fractions is partially counteracted by the larger size material having a greater probability of breakage (14).

It appears unlikely that shape factor is a function of the degree of grinding (at least, over the size range investigated). If it were, the factor

would be expected to vary for different sized fractions because the finer fractions contain material which has been broken several times. It is possible, however, that the shape factor varies from one type of grinding process to another.

An examination of Seyler's coal chart (15) indicates that over most of the coal range, an increase of 1% in the hydrogen content of a coal has an equivalent effect on the volatile matter (dry ash free) of about an 8.5% decrease in carbon content, the percentage being expressed on Parr's basis (16). Therefore, it is proposed that the rank of a coal be expressed by

$$\text{Rank Index} = \%\,C \text{ (Parr's basis)} - 8.5\%\,H \text{ (Parr's basis)} \qquad (9)$$

When the volatile matter contents of coals are plotted against this rank index, the points have a considerably reduced scatter about the mean line over that when percentage carbon content alone is used (17). This is also true when grindability indices are plotted against the proposed index as shown in Figure 4 (16). Figure 4 also shows the shape factors of the four coals tested as a function of the rank index. Although definite conclusions cannot be drawn from four results, it seems probable that the variation in shape factor has a similar relation to the rank of the coals as that found for grindability indices.

The geometric or hydrodynamic area obtained with the liquid permeability apparatus should not be a function of the chemical nature or roughness of the particle surface since the resistance to flow is due to the internal friction of the liquid. Also the mean free path of the liquid molecules is not great enough compared to the flow paths for the phenomena of slip to occur. Consequently, the variation of shape factor implies that coals fracture to different mean shapes depending on their rank. A high shape factor means that the particle is flaky, while shape factors approaching the value of six imply that the particles tend towards spherical or cubical shapes. Thus, on the basis of our limited results, anthracites and low rank sub-bituminous coals have more flaky particles, while the more easily broken bituminous coals tend to have more rectangular shaped breakage products.

It is possible that shape factors also depend to a considerable extent on the petrographic constituents of the coals, since it is known that different macerals have conchoidal, splintery, or irregular breakage (18).

CONCLUSIONS

The geometric-surface-area-to-volume shape factor was found to be constant for a given coal over the size range investigated, approximately 40 to 600 microns. It seems likely that the shape factors of coals are related to their rank in a similar manner to that of their grindability indices. Shape factors were found to vary from 7.2 for a medium rank bituminous coal (17.9% volatile matter, d.a.f.) to 9.4 for a high rank anthracite (4.5%) and 9.6 for a low rank sub-bituminous coal (42.4%).

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TABLE 1

ANALYSES OF COALS USED

Coal	B-19447	B-17790	B-19426	St. Nicholas Anthracite
Constituent	As used,%	As used,%	As used,%	As used,%
Moisture	1.5	0.8	0.5	1.6
Ash	16.5	7.8	14.5	9.3
Carbon	65.5(83.5)*	78.8(87.6)*	75.2(90.6)*	84.2(95.5)*
Hydrogen	4.7(5.9)*	4.8(5.1)*	3.9(4.5)*	2.4(2.2)*
Nitrogen	1.1	1.5	1.5	0.85
Sulfur	4.5	1.6	1.8	0.5
Oxygen (by difference)	6.2	4.7	2.6	1.1
Volatile Matter (D.A.F.)	42.4	29.2	17.9	4.5

*Parr's basis

TABLE 2

SIZE-WEIGHT DISTRIBUTIONS AND HYDRODYNAMIC SURFACE AREAS OF SIEVE FRACTIONS FOR COALS GROUND ACCORDING TO THE STANDARD HARDGROVE TEST

Sieve Range U. S. Standard Mesh	B-19447		B-17990		B-19426		S.N.A.	
	p*	S_o**	p	S_o	p	S_o	p	S_o
16 x 30	100		100		100		100	
30 x 35	64.25	178	79.97	138	80.37	132	75.26	166
35 x 50	54.05	235	71.78	217	73.13	192	61.10	211
50 x 70	35.55	403	54.06	307	56.00	296	26.98	378
70 x 100	27.02	565	44.78	439	47.45	408	17.67	552
100 x 120	18.48	710	35.62	598	38.03	502	10.78	670
120 x 140	16.89	865	32.78	714	34.81	684	8.99	786
140 x 170	14.63	1001	28.78	948	31.22	744	7.23	1000
170 x 200	12.95	1171	25.95	1036	28.29	944	6.20	1156
200 x 230	11.17	1403	22.26	1235	24.69	1065	5.03	1398
230 x 325	9.82	1730	20.26	1635	22.48	1355	4.30	1717
Minus 325	7.94		11.14		16.29		3.20	
% Weight lost on grinding	0.66		0.72		0.83		(-0.1)	
Mean Hardgrove Index	52		93		99		30	

*p = % by weight below upper sieve size
**S_o = Surface area per unit volume of coal in size range given, $cm.^2/cm.^3$

TABLE 3

SURFACE AREA TO VOLUME SHAPE FACTORS FOR COALS
GROUND ACCORDING TO STANDARD HARDGROVE TEST

B-19447			B-17990			B-19426			St. Nicholas Anthracite		
μ *	$\frac{dS}{dp}$ **	k ***	μ	$\frac{dS}{dp}$	k	μ	$\frac{dS}{dp}$	k	μ	$\frac{dS}{dp}$	k
47.5	1421	9.6	44	1420	7.9	44	1240	7.1	47.5	1340	9.6
62	1124	9.6	62	1040	8.1	62	900	7.3	62	995	9.4
74	935	9.5	74	850	8.0	74	809	7.3	74	850	9.6
88	782	9.5	88	726	8.0	88	632	7.3	88	690	9.3
105	678	9.8	105	610	8.2	105	535	7.4	105	591	9.5
125	565	9.7	125	480	7.8	125	486	7.2	125	475	9.2
138	497	9.6	149	396	7.9	149	357	7.1	149	406	9.4
220	338	9.8	220	268	8.0	210	250	7.2	210	286	9.4
297	230	9.4	297	197	8.0	297	173	7.1	297	201	9.3
500	140	9.6	500	128	8.3	500	100	7.1	500	113	8.9
590	118	9.6	590	104	7.9	590	87.7	7.35	590	101	9.3
Mean k		9.6			8.0			7.2			9.3

* μ is sieve size in microns

** $\frac{dS}{dp}$ is the specific surface area in $cm.^2$ per g. at size μ

*** k is volume-to-surface area shape factor

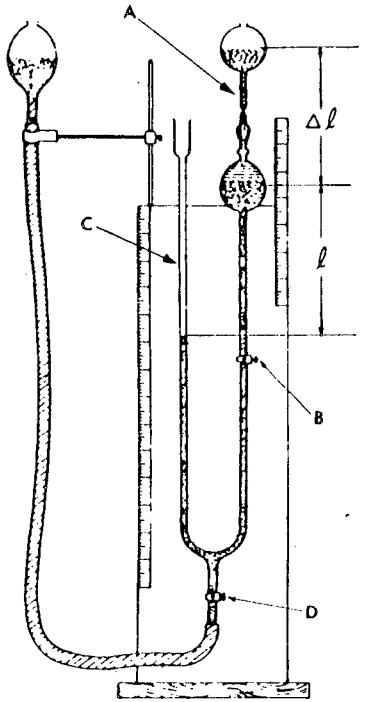

Fig. 1. DIAGRAM OF PERMEABILITY APPARATUS.

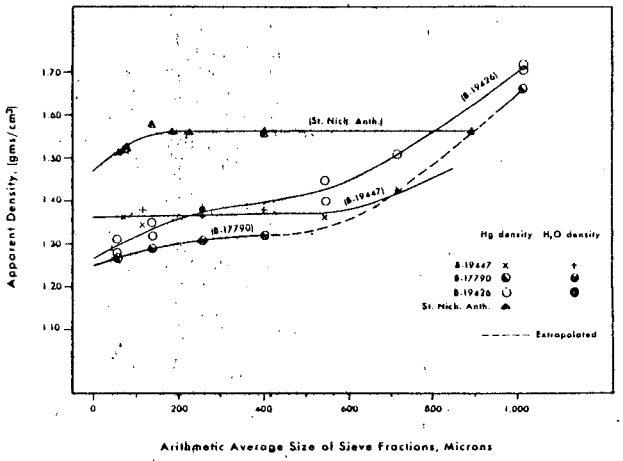

Fig. 2. APPARENT DENSITIES OF SIEVE FRACTIONS OF COALS GROUND ACCORDING TO STANDARD HARDGROVE TEST.

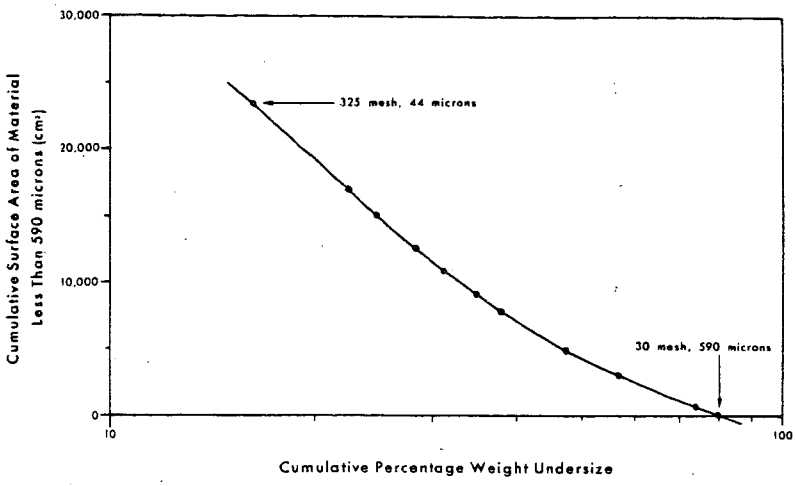

Fig. 3. CUMULATIVE SURFACE AREA AGAINST PERCENTAGE UNDERSIZE FOR COAL B-19426 GROUND ACCORDING TO STANDARD HARDGROVE TEST.

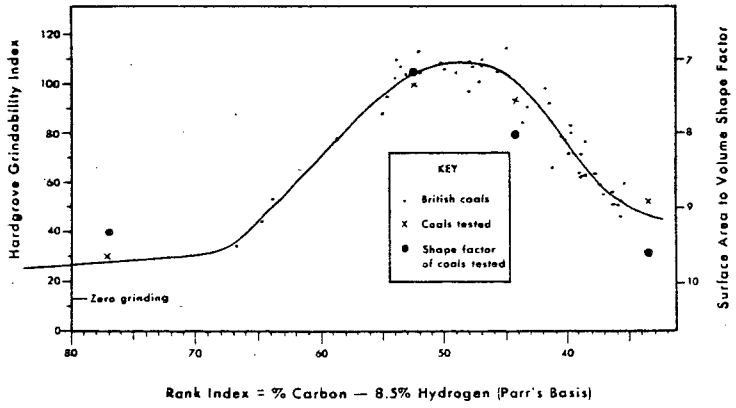

Fig. 4. GRINDABILITY INDEX AND SHAPE FACTOR AS A FUNCTION OF COAL RANK.